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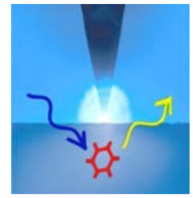

NEW INSTRUMENTATION FOR NANOSCALE SUBSURFACE SPECTROSCOPY AND IMAGING

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1. PROGRAM OBJECTIVE:

The objective of this MURI project was the development of techniques for non-destructive, chemically specific, three-dimensional nanoscale characterization of subsurface structures. The MURI team explored different measurement modalities (near-field spectroscopy, tunneling microscopy, fluorescence self-interference, and solid immersion lens microscopy) with the goal to combine them in a single measurement platform. A central goal was to combine the high chemical specificity provided by optical spectroscopy (e.g. IR absorption, Raman scattering) with near-field detection based on optical antennas (e.g. metal tips, particles). Operating in a transparent frequency-window made it possible to perform high-resolution chemical imaging of subsurface structures. The microwave regime has been explored for the detection of electron spins and dopants. Furthermore, using solid immersion lenses and fluorescence self-interference the team developed subsurface imaging of features buried at a distance of many wavelengths. The experimental effort was guided by theoretical studies aimed at the three-dimensional reconstruction of nanoscale objects with unprecedented resolution.

2. SCIENTIFIC APPROACH:

Te determination of chemical composition and material properties is accomplished by near-field microwave, infrared, fluorescence and Raman spectroscopy. The three-dimensional structure is reconstructed using near-field tomography and novel theoretical inversion algorithms. This MURI program was subdivided into four tasks as schematically represented by Figure 1.

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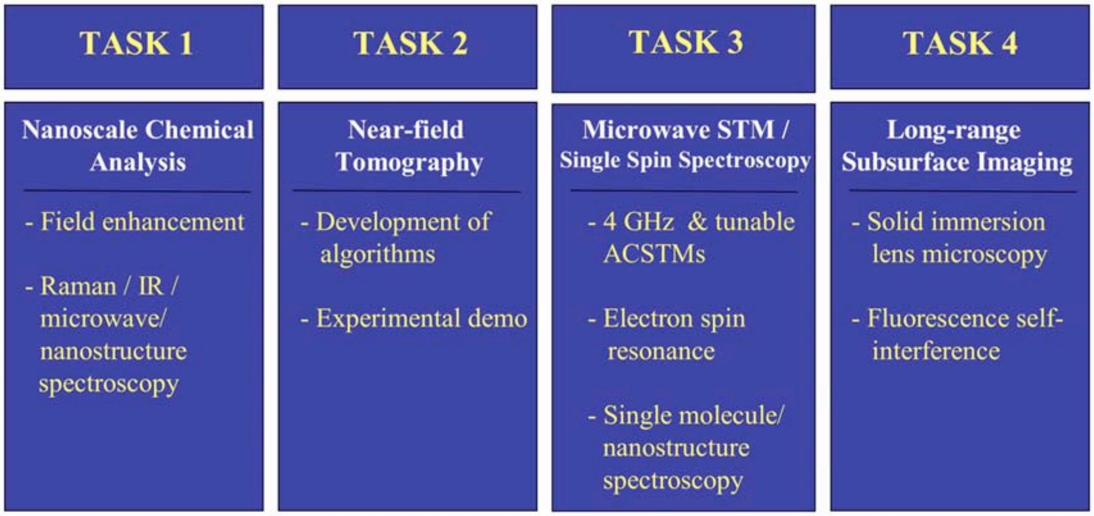

Figure 1: Structure of MURI program. The four tasks are defined according to the team members' expertise: Task 1 (Novotny / Stranick). Task 2 (Carney), Task 3 (Weiss / Kelly), and Task 4 (Goldberg / Unlu).

In the initial period of the project each task was aimed at the independent development and refinement of techniques in the laboratories of the participating team members. Test samples have been developed to demonstrate the feasibility of different measurement modalities, to assess their limitations, and to provide comparison with other, existing techniques. In funding year 3 the team began to converge the different efforts and started to plan the integration of different measurement modalities into a single measurement platform. The coordination between the different tasks was facilitated by an administrator (Barbara Schirmer, schirmer@optics.rochester.edu) who took care of maintaining an informative website (http://www.nano-optics.org/muri03). The website provided an efficient coordination of the MURI project, ensured a high visibility of the program, and helped to disseminate information, results and progress.

In funding year three and five, the MURI team organized dedicated workshops that were attended by all team members and by invited guests (http://www.nano-optics.org/muri03/workshop%2005). These meetings served as a platform to discuss the newest results, to develop future strategies, and to expose involved students to all aspects of this MURI program. Guests included Michael Burns from FEI company, Rainer Hillenbrand from Max Planck Institute, Germany, Michael Beversluis and Jeroen Schoenmaker from NIST, Gaithersburg, John Schotland from UPenn, Philadelphia, Stephen Ippolito from IBM Yorktown, NY, Anna Swan from Boston University, and George Lengel from RHK Technology, Troy, MI. Several ideas were born during these workshops and were later pursued by the team. For example, Rainer Hillenbrand (Max Planck Institute) came to spend his sabbatical at the Institute of Optics, University of Rochester, where he worked with Novotny's group on an instrument for simultaneous amplitude, phase, and polarization measurements of nanoplasmonic samples.

The MURI project proceeded in line with the original statement of work. The project resulted in the development of dedicated near-field measurement platforms. One of these instruments is being further developed in a collaboration between NIST (Stranick) and the

University of Rochester (Novotny). Another instrument platform is being pursued in an STTR project between RHK Technology and the Novotny group. This STTR project will commercialize the technology developed under the MURI program, thereby making it widely applicable for materials research.

3. ACCOMPLISHMENTS:

The MURI team has achieved various important milestones. The list below summarizes some of the highlights.

- Using near-field Raman scattering, Novotny's group has demonstrated chemically specific imaging with a record spatial resolution of 10 nm. These results were listed in the 2005 Guinness Book of World Records. For the first time, the team has also demonstrated the localization of defects and dopants in single-walled carbon nanotubes using near-field Raman scattering. The same technique was applied to image subsurface features buried by a layer of 5-10 nm of SiO_2 for acquiring near-field absorption spectra of individual proteins.
- Near-field inverse scattering and power extinction tomography developed by Carney has been demonstrated experimentally for the first time using photon scanning tunneling microscopy. The experiments established that not only the geometrical shape of an object can be reconstructed but also its susceptibility distribution.
- For the first time, the 'numerical aperture increasing lens (NAIL)' technique was applied to high-resolution light injection microscopy, an imaging modality commonly used for failure analysis by the semiconductor industry. Unlu and collaborators demonstrated a considerable improvement of the state-of-the art resolution and collection efficiency of through-substrate imaging of integrated circuits. To translate the imaging microlens on the surface of the sample a new slip-stick method has been developed and implemented.
- Weiss and Kelly achieved the first STM images of subsurface hydrides in a palladium surface. They were able to create subsurface hydrides as a reagent and they determined their electronic and structural properties. Simultaneously, they demonstrated manipulation of subsurface hydrogen atoms. Using microwave STM they achieved the first atomic-scale images of carrier concentrations in conducting polymers.
- Goldberg used fluorescence interference microscopy to achieve first measurements of DNA conformations relevant for micro-array technology. He has developed and implemented '4-π interferometric microscopy' and applied the technique to image biologically relevant systems such as *Shigella*. Several new techniques have been invented to improve 3D confocal imaging.

4. PUBLICATIONS:

Since this MURI has been launched the MURI team published 90 peer-reviewed publications in major research journals. A complete list is provided below. Four patent applications have been filed and more than 80 presentations have been given at conferences and workshops. It is important to emphasize that the MURI team members were organizers of more than 20 symposia, conferences, or workshops. This activity provided exposure to very large student, scientific, and industrial audience across physics, chemistry, biology, and engineering.

5. IMPACT:

The MURI team has achieved many firsts and set new benchmarks for high-resolution, chemically-specific imaging. That the MURI team members are leaders in this important research field is exemplified by the participation of Stranick and Weiss on the National Academy of Sciences Panel on “Chemical Imaging” (2005), and the participation of Novotny and Goldberg on the panel of the National Nanotechnology Initiative on "Instrumentation and Metrology for Nanotechnology'’ (Grand Challenge Workshop, 2004). In collaboration with IBM, the NAIL technique developed by Unlu has been implemented for the inspection and failure analysis of semiconductor integrated circuits. The NAIL technique surpassed the state-of-the-art imaging capabilities of the semiconductor industry and set new standard for optical resolution in ‘through-substrate’ imaging. The technology has been licensed to Hamamatsu and we worked with FEI company to fabricate lenses on the back side of Si substrates using their laser assisted etching technique. Weiss has demonstrated the ability to quantify dopant concentrations in semiconductors with ultrahigh resolution and is collaborating with leading semiconductor industries (Sematech, Intel, AMD, Texas Instruments). Finally, the high resolution images acquired with the near-field Raman microscope developed by Novotny have been featured in the Guinness Book of World Records. This listing provides a high visibility to our MURI effort as is evidenced by the many daily hits of the MURI webpage. Nanoscale Raman microscopy/spectroscopy is not only suitable for the structural analysis of nanoscale materials but could also play a significant role for the characterization of nanocomposite drugs. The latter are being increasingly developed by the pharmaceutical industries for reasons of solubility and drug uptake by the human body. However, currently there are no chemically-sensitive methods to analyze the nanocomposite nature of these drugs. Near-field Raman microscopy holds also great promise for characterizing local stress in semiconductor nanostructures. In fact, one of the big metrological challenges of the semiconductor industry is the characterization and control of stress. As devices become smaller and smaller local stress becomes a dominating factor for device performance and reliability.

6. TECHNOLOGY TRANSITION/TRANSFER:

The MURI team collaborated closely with industrial partners on different aspects of nanoscale metrology and characterization. The team has filed several disclosures and patent applications, and licensed NAIL technology to Hamamatsu (Japan). The team works with RHK Technology on the design of an instrument platform and on extended data acquisition and control capabilities (this collaboration is currently supported by an STTR Phase I grant). Among the industrial collaborators were: IBM, FEI, RHK, Sematech, SRC, AMD, Intel, and Texas Instruments. The team also established collaborations with government labs such as NASA (David Fischer), Argonne Nat’l Lab (Gary Wiederrecht), and with diverse people at NIST, Gaithersburg. The technology developed in this MURI program is critical for the chemically-specific characterization of a wide range of materials of importance to DoD. Nanoscale chemically-specific metrology is also critical for high-density electronics, bio-inspired devices, highthroughput experimentation (e.g. forensic sciences), nanocomposite materials (e.g. radar absorbing materials), single-site catalysis (e.g. chemical and energy conversion applications), and even actinide nanomaterials (e.g. generation of nuclear devices).

7. DESCRIPTION OF RECENT PROGRESS:

Here we summarize some of the most recent research progress. Descriptions of previous research accomplishments are detailed in earlier reports and can be downloaded from the MURI website (www.nano-optics.org/muri03).

We have developed a measurement platform to characterize and control optical fields on the nanometer scale, - a central theme of plasmonics and nanophotonics. Methods for characterizing localized optical field distributions are necessary to validate theoretical predictions, to test nanofabrication procedures, and to provide feedback for design improvements. Typical methods of probing near fields (e.g. single molecule fluorescence and near-field microscopy) cannot probe both the complex-valued and vectorial nature of the field distributions. We demonstrated that a nanoparticle probe with isotropic polarizability in combination with polarization control of excitation and detection beams provides access to this information through the interaction tensor. For a sample consisting of a single nanoparticle we showed that the recorded images correspond to maps of the local Green's function tensor elements that couple the probe and sample. The tensorial mapping of interacting nanoparticles is of interest for optical sensing, optical antennas, surface enhanced Raman scattering, nonlinear optics, and molecular rulers.

A sketch of the measurement platform is shown in Fig. 1. The probe-sample region is illuminated with a field $\mathbf{E}^i$, which interacts with the probe and sample, and is then scattered away

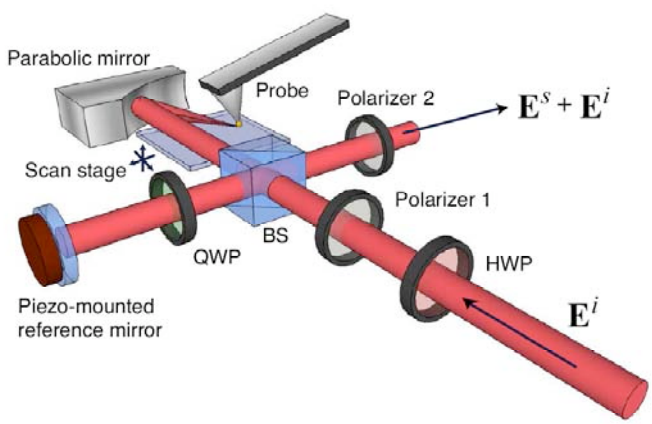

Fig. 1: Schematic of the measurement platform, consisting of an atomic force microscope and a phase-shifting Michelson interferometer, where a combination of waveplates and polarizers allow control of incident and detected polarizations. The half wave plate and polarizer 1 select the incident polarization, while the quarter wave plate and polarizer 2 select the detected polarization. At each pixel, the piezo-mounted reference mirror is translated, resulting in several measurements with different phase offsets. From this, the amplitude and phase of the signal are determined.

and collected interferometrically. The probe-sample distance is modulated at the frequency Ω and the detected signal is demodulated at 2Ω. This has the effect of suppressing signals that vary slowly in probe-sample separation, usually defined as background. Since it is necessary to decouple amplitude and phase measured in the interferometric signal, we used phase-shifting interferometry developed previously in this MURI program (see Ref. [6] at the end of the document). At each image pixel, the reference mirror is translated in incremental steps Δx_i from a start position to an end position. Each step Δx_i gives rise to a phase offset δ_i, allowing the interferometric signal at each mirror position (denoted by subscript 'i') to be written

$$I_i = A + R \cos (\Phi + \delta_i) \quad (1)$$

where A is a non-interferometric offset, R is proportional to the amplitude of the scattered field, and Φ is the phase of the scattered field relative to a constant. A least-squares fitting algorithm then determines A, R, and Φ for each pixel. In practice, $A \approx 0$ after demodulation. Polarizers and waveplates are used to control the incident and reference polarizations. Light from an 831nm diode laser passes through a half-wave plate and then through a linear polarizer, allowing rotation of linear polarization incident on the sample. A quarter waveplate in the reference arm rotates the reference polarization when double-passed. Finally, the combined beams pass through a linear polarizer, which selects the polarization of the measured signal. With these elements, combinations of incident and scattered fields from the probe-sample region, notably P and S polarization, are analyzed. The experiment is only weakly sensitive to the angle of the waveplates, which control the light intensity transmitted by the polarizers.

During a scan, the incident and scattered fields are analyzed with polarizers set to either S polarization or P polarization. This renders four distinct polarization measurements, namely SS, SP, PS, and PP (first index refers to incident light and second index to scattered light). We note that the background is in general unrelated to the non-interferometric offset A from Eq.(1), but instead manifests in the measurement of R and Φ. The field scattered from the probe-sample region (E_{scat}), is a superposition of fields scattered from the probe and sample. Theoretical studies performed under this MURI (see Ref. [35] at the end of the document) that this field can be generally represented by

$$\mathbf{E}_{scat} = \Sigma\, (S + T)^n \cdot \mathbf{E}_i \quad (2)$$

where the index n denotes the scattering order and T and S operate on the incident field, $\mathbf{E}_i$. They represent single scattering events from the probe (T) and sample (S). Our experimental situation simplifies Eq. (2) considerably. The probe and sample are assumed to be identical weakly-scattering point dipoles located at $\mathbf{r}_T$ and $\mathbf{r}_S$. Assuming that the scattering series in Eq. (2) converges, we obtain a first-order approximation by truncating the series to $n \leq 2$, which allows us to represent the detected field as

$$\mathbf{E}_{scat} = (S + T + SS + TT + ST + TS) \cdot \mathbf{E}_i \quad (3)$$

The quantity in parentheses is the (truncated) optical interaction tensor between the probe and sample, and the incident and detected field polarization states can be used to select combinations of its elements. Demodulation at the 2nd harmonic of the probe-sample modulation frequency suppresses the S, T, SS, and TT terms, leaving only ST and TS terms, which can be determined by polarization sensitive measurements.

To demonstrate our ability to measure the full complex and vectorial nature of optical near-fields we have chosen a test sample with discrete gold nanoparticles. For pairs of single gold nanoparticles, the ST and TS operators can be represented by the dydic Green's function G($\mathbf{r}_T$, $\mathbf{r}_S$). Fig. 2 shows the amplitude and phase images for all four combinations of incident and detec-

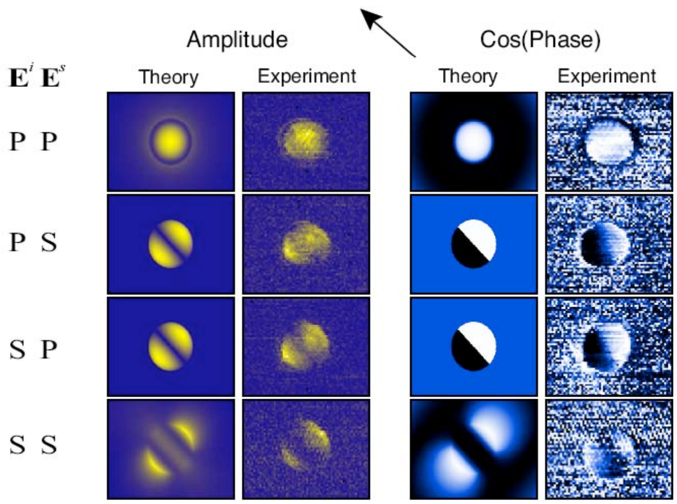

Fig. 2: Amplitude and phase images of a single 100nm diameter gold nanoparticle for various polarization configurations. The images correspond to different components of the interaction tensor G($\mathbf{r}_T$, $\mathbf{r}_S$)*, where* $\mathbf{r}_T$ *and* $\mathbf{r}_S$ *denote the coordinates of the two gold nanoparticles. The arrow indicates the direction of incident light.*

ted polarizations along with corresponding images of the Green's function tensor elements. Several qualitative features of the experimental and simulated data are evident. In the PP case, a bright center and a faint ring are visible in the amplitude image, and the phase is nearly constant across the particle. The experimental SP and PS cases are similar, and identical in theory. They show two bright lobes in amplitude, but the scans exhibit an asymmetry in their brightness, most likely due to slight mixing of the S and P polarization states by the parabolic mirror. Such a mixing would cause the dipoles of probe and sample to be slightly misaligned with the scan plane. Though two bright lobes are seen in all amplitude patterns except PP, the phase images clearly distinguish between the SP/PS cases and the SS case. There is a sharp jump of ≈ 180 degrees in the SP and PS phases. The SS phase has a complicated pattern, and the associated low signal makes a comparison difficult, but the two characteristic bright lobes are present on the particle, along with a dark central stripe. The noise in the phase images is due to the vanishing field strength for larger particle-particle separations.

We have chosen two gold nanoparticles as a simple system in which tensorial information about amplitude and phase is important and easily distinguished. In addition to this, the gold nanoparticle dimer system has applications in optical sensing, optical antennas, surface enhanced Raman scattering, nonlinear optics, and molecular rulers, for which the interaction tensor between the particles is of interest. As opposed to standard tip-based near field microscopy, our nanoparticle approach makes it possible to retrieve all relevant components of the interaction tensor. Polarization sensitivity in near-field optics provides insight into the tensor nature of the probe-sample interaction, and in special cases helps to isolate individual Green's function elements. This additional degree of freedom, along with well-defined probe geometries, such as gold nanoparticles used in this work, are additional tools for fundamental investigation of plasmonic and photonic systems.

It is planned that the tensorial measurement modality described above will be fully implemented in a commercial instrument platform. The development of this platform is the objective of an ongoing STTR grant with RHK Technology.

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